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CHEMICAL DIVERSITY OF TALCS IN RELATION TO THEIR ORIGIN

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ABSTRACT

Apart from main constituents, chemical compositions of talc show various elements, such as Fe, Al, Cr, Ni, Mn, alkalies and fluorine. Several analyses of talcs from Egypt (El-Sharkawy 2000) show the rectilinear negative correlation of Si and Fe^{3+} suggesting simultaneous substitution of Fe^{3+} both in tetrahedra and in octahedra according to the mechanism $MgSi \leftrightarrow Fe^{3+}Fe^{3+}$. This was veryfied in talks deficient in Si and rich in Fe. Essentially better calculated formulas and agreement of the layer charge with the theoretical value 22 were achieved after recalculation of Fe oxidation state according to the above mentioned scheme. The highest Fe^{3+} contents demonstrate talcs from low-tmperature crystallization environments. The likely explanation is interaction with pore fluids.

Chemical compositions of octahedral sheet, plotted in the ternary diagram Mg-Al-Fe+Mn+Ni+Zn, demonstrate a quite regular grouping of projection points into 3 fields: I. near-stoichiometric talcs around 100 % Mg, II. talcs with elevated Fe+Mn+Ni+Zn contents and III. Mg-Al talcs. Samples representing all genetic types gather in the first field, in the second dominate talcs from high-pressure and ultramafic environments, and in the third - dolomite-related talcs.

The positive correlation of tetrahedral Al and Na (R^2=0.71) suggests the substitution mechanism involving the tetrahedral sheet and interlayer space: $NaAl \leftrightarrow Si$. Much weaker correlation shows octahedral Al (R^2=0.50). Sodium content shows positive correlation both with temperature and pressure of talc crystallization. It is likely that high PT-conditions favour incorporating Na into the talc structure.

Key words: talc, crystal-chemistry, chemical composition

INTRODUCTION

The aim of this work was to summarize the available chemical analyses of talcs to derive possible crystal-chemical signals of their origin and structural features. The authors of the present study collected 121 chemical analyses of talc, both published and obtained in the own laboratory. Talc belongs to 2:1 layer silicates which have simple theoretical formula $Mg_3[(OH)_2|Si_4O_{10}]$. Nevertheless, it usually contains other chemical elements in minor amounts.

Talcs can be categorized according to the type of rocks in which is found, e.g. dolomites or serpentinites. Nevertheless, they also can be categorized regarding the mineral-forming process, for example metasomatic, hydrothermal, and the crystallization conditions, such as temperature or pressure. The chemical and structural parameters might be indicative of the origin and crystallization conditions of talcs. The pool of chemical analyses of talcs was divided into 5 genetic categories according to Evans and Guggenheim (1988) with some modifications. Because the genetic information provided together with published chemical data are often very scarce, the modifications consisted in widening the original genetic categories of Evans and Guggenheim (1988) were needed in order to fit as many cases as possible. However, the proposed categorization became not fully consistent, and ascribing some talcs to the proposed classification was more or less ambiguous. Therefore, the results should be treated with caution. Accordingly, the proposed categories are:

1. "Ultramafic and metamorphic", comprising talcs associated with ultramafites, serpentinites and regionally metamorphosed rocks – U,
2. "Dolomitic" describes talcs associated with metadolomites and dolomites – D,
3. "Low-Temperature" - comprising talcs occurring in low-temperature environments, including hydrothermal and metasomatic – H,
4. "High-Pressure" talcs – P,
5. "Sedimentary". This category embraces talcs associated with rocks of clear sedimentary origin (excluding carbonates) – O.

The genetic data were complemented by crystallization temperatures when available in the source literature. In majority of cases, there was given in the literature one temperature of formation of mineral paragenesis (or assemblage), and a series of mineral compositions. However, not always the genetic category directly corresponds to the temperature of crystallization.

VALENCY OF Fe

Iron substitutes for magnesium in limited amounts due to structural limitations (e.g. Forbes, 1969), however, Fe-bearing talcs were reported from many localities. Both ferrous and ferric iron was encountered in talcs (e.g. Petit et al., 2004), the second one occupying both octahedral and tetrahedral structural positions. In majority of cases, the chemical composition of talc is measured by using techniques which do not provide information about the oxidation state of elements, such as electron microprobe or X-ray fluorescence. Therefore, the presence of iron in various oxidation stages makes proper calculation of chemical formula of talk on the basis of charge-balance difficult. The partitioning of total Fe into ferrous and ferric Fe may help in achievement of better agreement of layer

charge with a theoretical value of 22. Nevertheless, such purely arithmetical approach may be or may be not in agreement with the real behaviour of iron in talc.

A general insight into the scale of uncertainty in calculation of crystal-chemical formula of talc gives the calculation based on 7 cations. The measure of inconsistency with theoretical formula may be the magnitude of discrepancy between the calculated layer charge and the value 22. From 121 chemical analyses of talc, recalculated to the formulae based on 7 cations, the layer charge was <21.9 in 13 samples, >22.1 in 32 samples, and the remaining 78 had layer charge of intermediate values. Beside of other factors, these inconsistencies with the theoretical value 22 may stem from the different oxidation state of Fe than that which was assumed.

Direct measurements of the Fe oxidation state in talcs are rare. One of the authors, who reported the independently measured Fe^{3+}-contents in a suite of talcs from Egypt, is El-Sharkawy (2000). His analyses trace a negative linear trend of Fe^{3+}-content along with the Si-content. After removing of 2 projection points as outliers, the R^2 coefficient is equal 0.93 (Fig. 1) and the linear regression equation is Y=7.29-1.82*X. It is shown by the equation that the deficiency in Si of the magnitude *n* relative to 4 corresponds to Fe^{3+}-content of *2n* in rough estimate. The negative correlation of these two elements likely points to a mutual substitution of Si and Fe^{3+} in tetrahedral structural positions in talc. However, in talc the amount of Fe^{3+} is twice as big as the magnitude of this substitution. The likely explanation is the occurrence of a simultaneous combined substitution of ferric iron both in tetrahedral and octahedral layers $MgSi \leftrightarrow Fe^{3+}Fe^{3+}$. The proposed substitution assures the local charge balance which enhances the stability of the structure (Bailey, 1984).

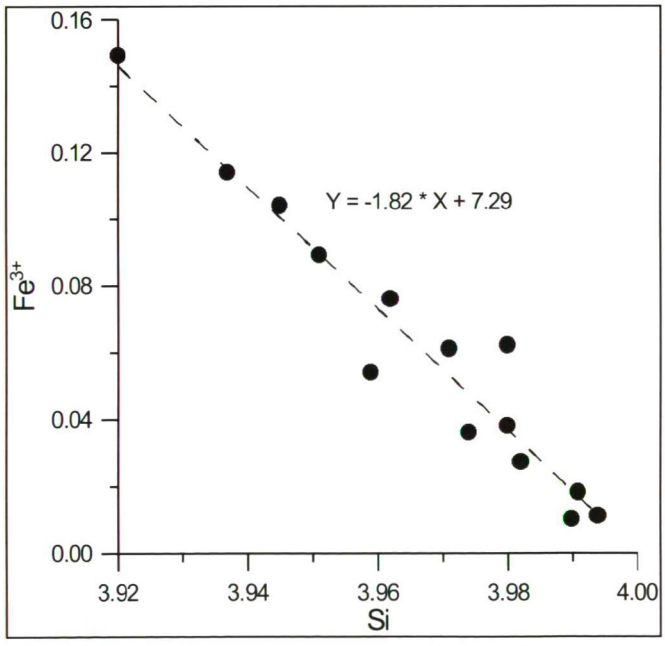

Fig. 1. Rectilinear correlation of Si and ferric iron contents in crystal-chemical formulae of talc from Egypt (El-Sharkawy 2000). The fitting line parameters are given.

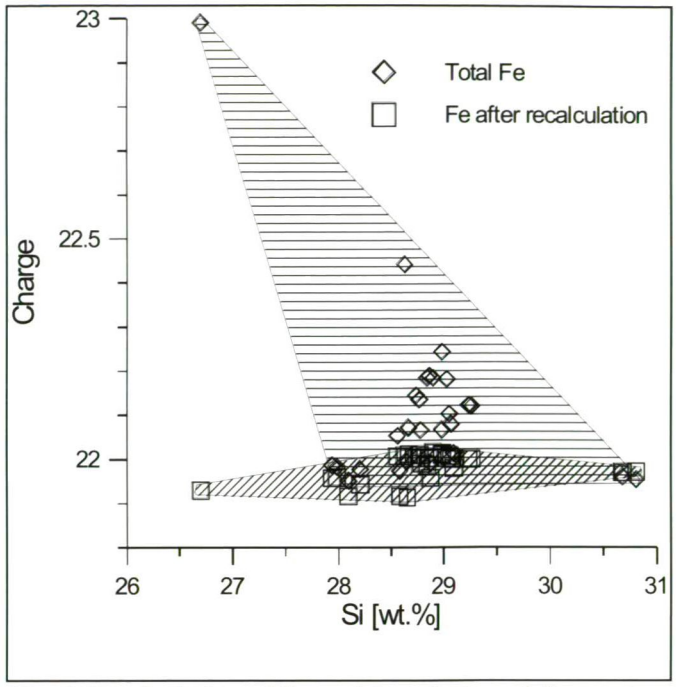

Fig. 2. Recalculation of Fe-total into ferric and ferrous iron according to scheme showed in the Fig. 1 leads to minimization of deviations of layer charge around the value 22, confirming the occurrence of the substitution $MgSi \leftrightarrow Fe^{3+}Fe^{3+}$.

In order to verify this hypothesis, the talc analyses with Fe given as Fe-total were again recalculated to chemical formulae on the basis of 7 cations, considering partitioning of total Fe to ferric and ferrous Fe there, where it was possible, according to the proposed scheme. The obtained layer charges much better approached the value 22 (Fig. 2). The mean layer charge in 27 analyses taken to this calculation including 12 analyses of El-Sharkawy (2000) changes from 22.17 to 21.98. Also, the variance expressed as the standard deviation decreased from 0.21 to 0.03. The obtained results proved that the proposed cations substitution occurs not only in talcs analyzed by El-Sharkawy (2000), but concerns other talcs deficient in Si and containing Fe.

The elevated ferric iron content preferably show talcs crystallized in lower temperatures (Fig. 3). The interaction with pore fluids rich in oxygen, for example with meteoric water in course of the talc crystallization, might be the possible explanation of this phenomenon.

After having distributed Fe according to the proposed scheme, the talc analyses were recalculated to chemical formulae based on 22 valencies. Unlike the calculation based on 7 cations, the present one considers the possible vacant sites in octahedral positions.

In the stoichiometric talcs, the sum of tetrahedral (*t*) and octahedral (*o*) charges are equal 16 and 6, respectively. Accordingly, the value $x = |10 - (o - t)|$ may be the measure of the goodness of a talc analysis in the neutral-lattice talc structure. Among the studied talcs, 80 % have the value $x <$ 0.3. The analyses showing high x-values seemed to be erroneous and were rejected. Beside of Si, Mg and O, the chemical analyses showed minor contents of Al^{3+}, Fe^{2+}, Fe^{3+},

Mn^{2+}, Ti^{4+}, Cr^{3+}, Ca^{2+}, K^{+}, Na^{+}, V^{5+}, Zn^{2+}, Ni^{2+}, Co^{2+}, Cu^{2+} as well as F^{-} and Cl^{-} as anions. The divalent cations were assumed to substitute for Mg, Al for Si in tetrahedra, and excessive Al substitutes for Mg in octahedra as well as Ti and V.

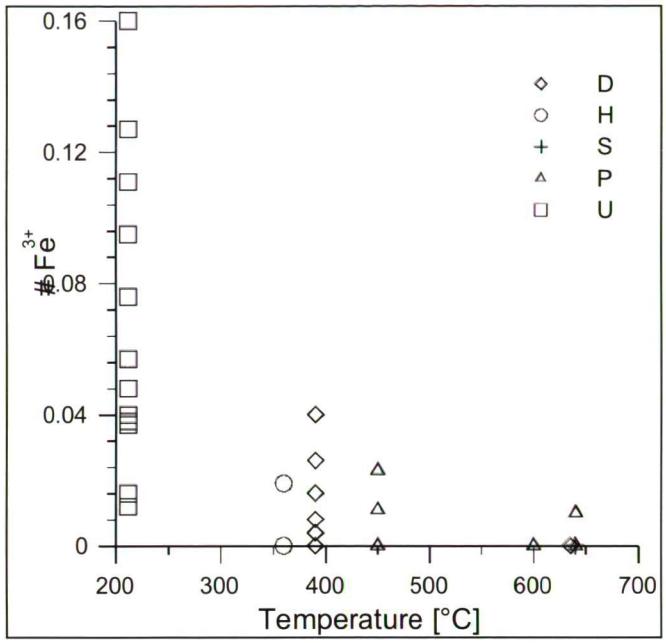

Fig. 3. The negative correlation of ferric iron and temperature of crystallization. The possible explanation is the oxidizing role of pore fluids. Symbols depict talc categories according to the text: *diamonds* - Dolomitic, *circles* - Low-Temperature, *plus-signs* - Sedimentary, *triangles* - High-Pressure, *squares* - Ultramaphic and Metamorphic.

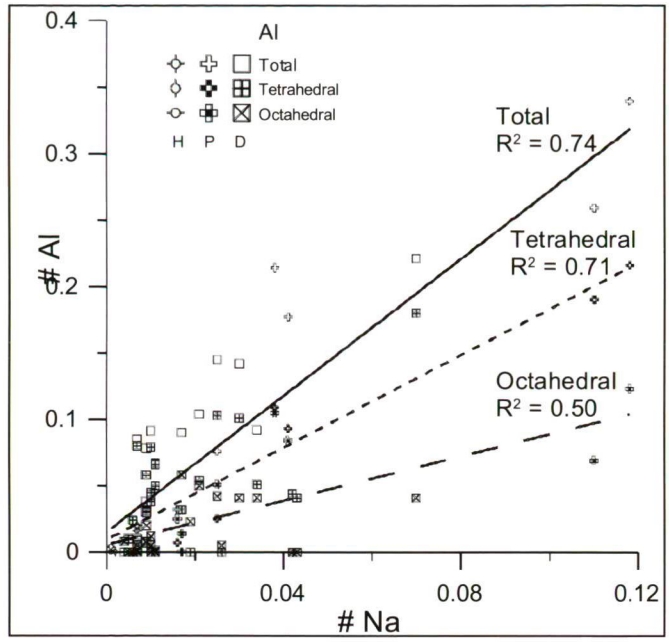

Fig. 4. Positive correlations of Al and Na suggesting the occurrence of the substitution mechanism NaAl↔Si. Sodium-bearing talcs were only found in H, P and D talc categories. See text for details.

SODIUM AND ALUMINUM

The elevated value of *x* may also stem from the presence of elements not atributed to either tetrahedral or octahedral sites. Location of Ca, Na and K cations is unclear. Unlike micas, due to the shift of adjacent 2:1 layers ensuring minimization of Si^{4+}-Si^{4+} repulsive forces (Zvyagin et. al., 1969), the talc structure seems not to have enough space in interlayer to accommodate larger interlayer cations. Nevertheless, Schreyer and Abraham (1976) postulated that Na^{+} could fit inside some of the hexagonal rings of the tetrahedral layers of talc. Recent modelling of Wang *et al.* (2004) showed, that limited number of water molecules can enter the interlayer space of talc-like 10-Å phase at elevated pressures and temperatures. The sizes of Na^{+}-cation and water molecule are very similar to each other. Therefore, it is supposed that a limited number of Na^{+}-cations might be inserted into the interlayer space in the talc structure at high pressures and temperatures, however, it should be complemented by cation substitutions compensating the layer charge.

The collected chemical data proved that the talc analyses showing presence of Na usually also contain Al, and these 2 variables are positively linearly correlated (Fig. 4). The coefficient R^2 has higher value for correlation Na with Al^{IV} (0.71) than with Al^{VI} (0.50), suggesting the substitution mechanism involving the tetrahedral sheet and interlayer space: NaAl↔Si. Such substitution in talcs was suggested also by Abraham and Schreyer (1975). The lack of a similar correlation with potassium may stem from the larger dimensions of K^{+}-cation preventing it from entering into interlayer space limited in size due to the above mentioned shifts of adjacent 2:1 layers. Highest Al and Na contents show talcs from high-pressure environments (Fig. 4), these talcs also show high temperature of crystallization (Fig. 5).

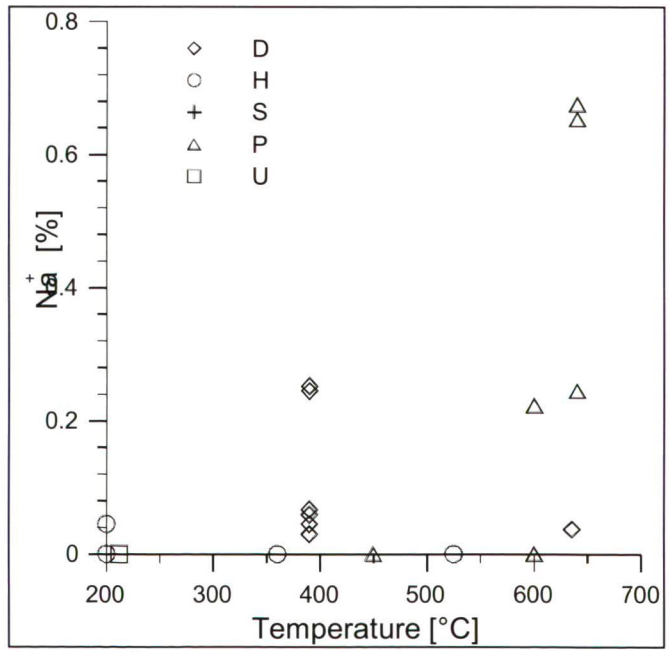

Fig. 5. Sodium content increases along with the increasing temperature of crystallization. Not all talc categories are represented due to the scarcity of available data. Symbols same as in Fig. 3.

The mechanism of sodium entering the talc structure is not clear. Fumagalli et al. (2001) suggest that the "10-Å phase" can act as the "high-pressure molecular sieve". Therefore it is supposed, that sodium dissolved in aqueous fluids might be incorporated in talc "by chance" together with water molecules.

Nevertheless, some talcs containing Na and Al linearly correlated with each other, studied by Schreyer *et.al.* (1980), showed broad basal reflections in XRD-patterns suggesting that it might represent a disordered mixed layer phase of talc and sodium phlogopite.

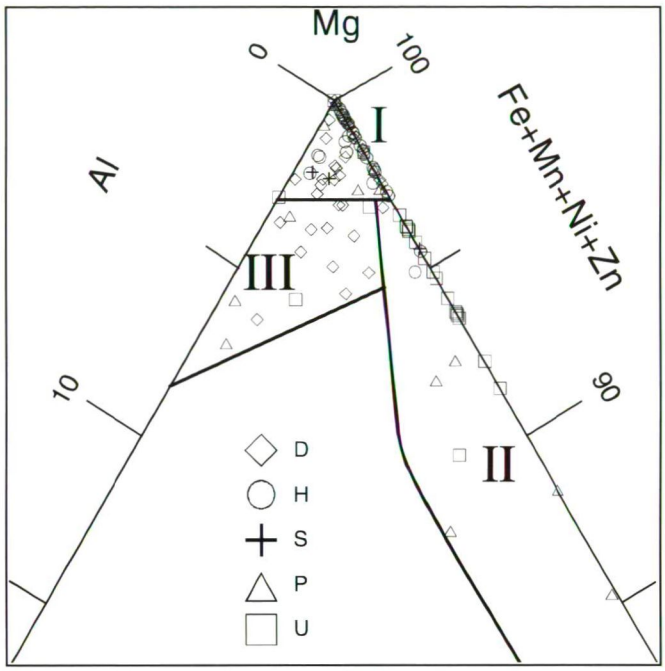

Fig. 6. Ternary diagram Mg-Al-Fe(+others) demonstrating chemical variability of the octahedral sheet in talcs. The configuration of projection points allows to distinguish 3 projection fields. Symbols same as in Fig. 3. See details in text.

Composition of the octahedral sheet

The chemical composition of the octahedral sheet of the studied talcs was projected in the ternary diagram Mg, Al and Fe+Mn+Ni+Zn+(Co+Cu) (Fig. 6). Both ferrous and ferric octahedral iron were considered. The majority of analyses plots near the Mg apex in the range 97-100% Mg showing near-stoichiometric compositions, and forming a densely populated projection field I. Remaining analyses plot in two distinct areas: near the Mg-Fe margin – field II, and analyses with higher Al-contents – field III. In the field I are projected analyses of all genetical types of talc, the field II mainly embraces the "ultramafic" and the "high-pressure" talcs while the field III – "dolomitic" talcs.

Preliminary studies on the nature of variability of degree of order of the crystal structure of talcs, manifested by the presence or lack of structural and super-structural reflections in XRD powder patterns, did not show any clear correlations with chemical parameters.

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